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Highly efficient and stable electrooxidation of methanol and ethanol on 3D Pt catalyst by thermal decomposition of In_2O_3 nanoshells

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ABSTRACT

In this paper In_2O_3 nanoshells have been synthesized via a facile hydrothermal approach. The nanoshells can be completely cracked into pony-size nanocubes by annealing, which are then used as a support of Pt catalyst for methanol and ethanol electrocatalytic oxidation. The prepared In_2O_3 and supported Pt catalysts ($\text{Pt}/\text{In}_2\text{O}_3$) were characterized by X-ray diffraction (XRD), energy dispersive X-ray spectroscopy (EDS), X-ray photoelectron spectroscopy (XPS), field effect scanning electron microscopy (FESEM), and transmission electron microscopy (TEM). Cyclic voltammetry (CV), linear sweep voltammetry (LSV), chronoamperometry and electrochemical impedance spectroscopy (EIS) were carried out, indicating the excellent catalytic performance for alcohol electrooxidation can be achieved on $\text{Pt}/\text{In}_2\text{O}_3$ nanocatalysts due to the multiple active sites, high conductivity and a mass of microchannels and micropores for reactant diffusions arising from 3D frame structures compared with that on the Pt/C catalysts.

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1. Introduction

Because of the rapid energy consumption and being exhausted on the traditional fossil fuel, energy crisis becomes more and more serious. Great attention has been paid the last decade to find new, green and sustainable energy sources. For powering portable electronic devices, fuel cell vehicles, and other devices, direct alcohol fuel cells (DAFCs) have been widely investigated [1–6]. DAFCs have more advantages as compared with hydrogen-based fuel cells, because the DAFCs have a lot of advantages such as low operating temperature, high energy density, and low pollution emission [7–11]. Both methanol and ethanol respectively have the considerable energy density of 702.32 and 1325.31 kJ/mol, but due to the lower toxicity, higher abundance, and wider convenience in industry, ethanol is the best choice [12–14]. For the realization of DAFCs application, the key factor is an effective catalyst. Among numerous electrocatalysts, Pt and Pt-based electrocatalysts are still the most effective catalysts for the electrooxidation of alcohol in DAFCs

[15–17]. However, the high cost and limited source of Pt are the blocks of commercialization of DAFCs [18]. As a result, reducing the use of noble metal Pt and improving the activity of electrocatalysts are great challenges. Now, there are two methods to solve the problems. The one approach is to use the Pt-based alloys and oxides and the other way is to efficiently utilize Pt by distributing limited Pt nanostructures on a novel support. Many scientists have concentrated on developing new anodic electrocatalysts that are the excellent supports for loading Pt nanoparticles. In the past few years, Pt-based nanocatalysts supported on carbon black have been widely used as anode electrode materials in DAFCs [19]. However, the presence of micropores (less than 1 nm in diameter) in carbon black limits its use as a catalyst support because the catalytic nanoparticles get trapped in the micropores and become electrochemically inaccessible [20, 21]. Now, it is found that certain metal oxides, such as MoO_3 , SnO_2 , and WO_3 can promote the activity and CO-poisoning tolerance of Pt-based catalysts for alcohol electrooxidation through synergetic interaction with Pt [22]. The perfect dispersion of Pt nanoparticles and the stability of combination of Pt nanoparticles and substrates can improve effectively the electrocatalytic activity.

Different nanostructures, such as nanowires, nanotubes, nanorods and nanobelts have been employed widely in the field of electrocatalysis, because many novel physical and chemical

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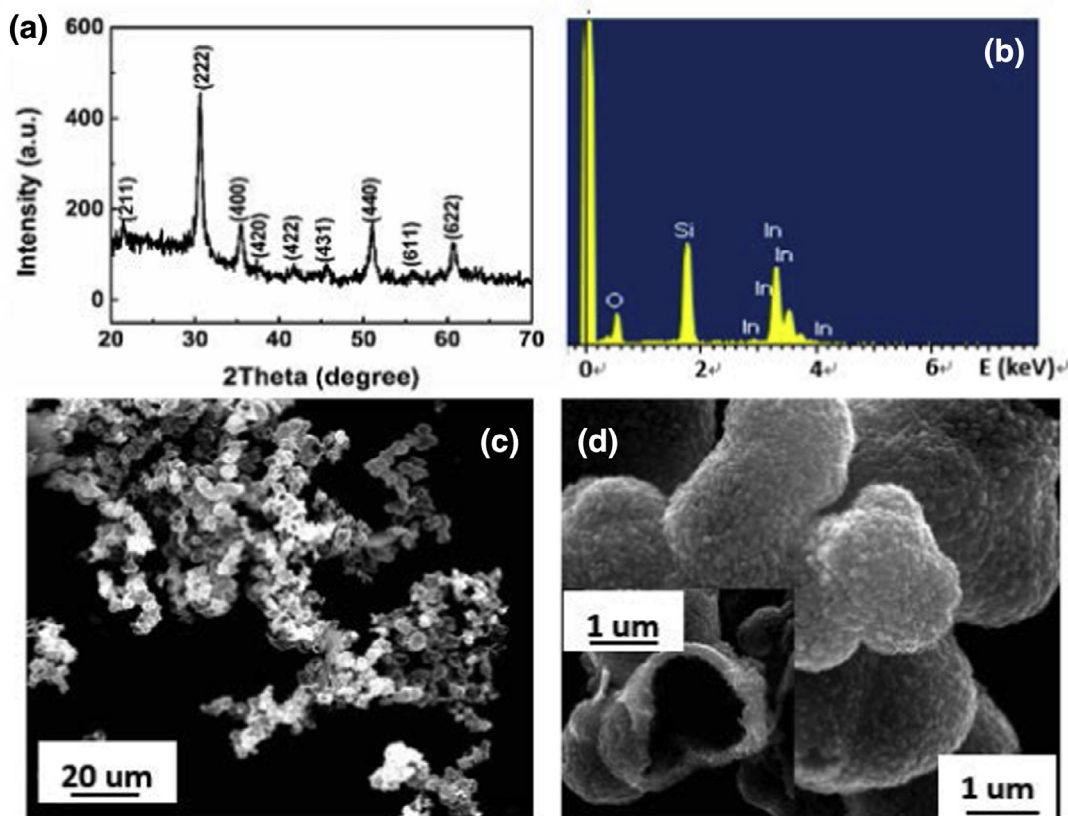


Fig. 1. (a) XRD pattern, (b) EDS result, (c, d and inset of d) FESEM images of the synthesized In_2O_3 nanocrystals.

properties can be delivered from this architecture. In_2O_3 is a wide band-gap (direct band gap about 3.6 eV) n-type semiconductor and has low resistivity as well as good catalytic activity [25]. However, although In_2O_3 is widely utilized in the field of gas sensors, there are few reports of In_2O_3 used as the substrate of Pt nanoparticles in DAFCS. Due to the large surface area, high electrical conductivity, effective mass transport and stable chemical property of In_2O_3 , it is necessary to investigate In_2O_3 nanostructures as supports to load Pt for electrocatalytic oxidation of alcohols.

Herein, the In_2O_3 has been synthesized by a hydrothermal method and utilized as the 3D support for Pt loading. Pt nanoparticles have been successfully coated on the In_2O_3 nanocubes to perform as catalyst for alcohol electrocatalytic oxidation. We have investigated systemically the performance of the $\text{Pt}/\text{In}_2\text{O}_3$ catalysts for methanol and ethanol electrooxidation in the acidic media by CV, LSV, chronoamperometry and EIS. By using the test methods, it is demonstrated that the $\text{Pt}/\text{In}_2\text{O}_3$ shows desirable electrochemical activity and stability.

2. Experimental

2.1. Chemicals

$\text{In}(\text{NO}_3)_3 \cdot 4.5\text{H}_2\text{O}$, DMF (dimethyl formamide), EGME (ethylene glycol monomethyl ether), $\text{H}_2\text{PtCl}_6 \cdot \text{H}_2\text{O}$, methanol and ethanol are purchased from Aladdin Industrial Corporation. However, nafion and silver paste are obtained from Sigma–Aldrich and SPI Suppliers. Similarly carbon black with the type of Vulcan XC72R is acquired from CABOT Corporation. All these chemicals used here are analytically pure. Deionized water has also been used throughout.

2.2. Synthesis of $\text{Pt}/\text{In}_2\text{O}_3$

The In_2O_3 nanoshells are synthesized by a simple hydrothermal method. 1.15 g of $\text{In}(\text{NO}_3)_3 \cdot 4.5\text{H}_2\text{O}$ and 2 mL of DMF are dissolved in 30 mL of EGME under continuous magnetically stirring for 1 h. Then, the mixture was transferred into a 55 mL sealed Teflon-lined autoclave for heating at 200 °C. After reaction for 24 h, the autoclave was taken out and cooled down to room temperature [26]. Subsequently, the obtained sediment was washed with deionized water and ethanol. Followed by this it was then dried for further utilization.

To obtain $\text{Pt}/\text{In}_2\text{O}_3$, 0.04 g of dry In_2O_3 was mixed with 50 μL of 85 mM $\text{H}_2\text{PtCl}_6 \cdot \text{H}_2\text{O}$ solution. After ultrasonically being treated for 10 min, the $\text{H}_2\text{PtCl}_6 \cdot \text{H}_2\text{O}$ and In_2O_3 are mixed evenly, which is then calcined at 380 °C for 40 min to thermally decompose H_2PtCl_6 on In_2O_3 nanoframes. After being washed with deionized water and absolutely ethanol, the dark gray $\text{Pt}/\text{In}_2\text{O}_3$ nanostructures are collected.

The crystalline phase of the sample is identified by XRD. The morphology and the size of In_2O_3 and $\text{Pt}/\text{In}_2\text{O}_3$ are observed by FESEM and TEM. EDS and XPS are used to examine the chemical component.

2.3. Electrode fabrication and electrochemical measurements

The graphite electrode is rinsed with deionized water and washed ultrasonically with absolute ethanol. To fabricate the working electrode, a certain content of $\text{Pt}/\text{In}_2\text{O}_3$ nanocatalyst ethanol ink is painted on a graphite electrode with the window area (of about 0.2 cm^2). Then, 15 μL Nafion solution with the concentration of 0.5 wt% is dropped on the electrode surface to immobilize the catalysts and improve the anti-interference ability [27]. Pt loading on the electrode was 0.25 mg/cm^2 . For comparison, Pt supported

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